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LIGHT AMPLIFICATION AND LASER EMISSION FROM SPECIAL MICROMETRIC CONFINED SYSTEMS

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The mélange of light localization and laser emission is particularly fascinating due to the specific features of each distinctive lasing source, accompanied by exclusive emission properties of the localized modes. In this manuscript we demonstrate light amplification and random laser effects from special dye-doped confined micrometric systems. We physically characterize the particular sample configurations for low and high power pumping conditions from spectral point of view. Analysis of far field spatial emission confirms that light localization and random lasing occurs in our systems upon exceeding a certain input energy threshold. In addition, the presented small-size optical systems are also highly robust and prone for immediate exploitation in the micro technological arena.

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1. Introduction, experimental set-up and samples

Lasing phenomena and in particular random lasers have been a domain of intense theoretical and experimental studies in the last years. They are captivating examples of scientific topics that coalesce multiple scattering of light and optical amplification. Subsequent to the innovative work of Letokhov et al. from 1968 [1], various hosts such as semiconductor powders [2-4], silver nanopowders [5], polymers [6], liquid crystalline materials [7-10] and even human tissues [11] have been employed for engineering unique and exotic systems in obtaining light amplification and eventually lasing.

Specifically, in a random laser system, several discrete modes are optically amplified within the host medium and exhibit individual resonant wavelengths with the spectral interval being usually in the range of a few nanometers. The mélange of light localization and stimulated emission is remarkably alluring due to the specific properties of each distinctive lasing source, complemented by unique emission characteristics (given by localized modes in every sample). Many important research studies in the field [12-22] contributed enormously in achieving new perspectives in the aforementioned scientific areas. The opportunity to employ novel materials while allowing for original flexible designs in engineering compact and reliable laser sources represents nowadays a greatly motivating race.

In this manuscript, we show the design and fabrication of active dye-doped ionic liquid lasers at micrometric scale, we investigate the optical amplification mechanism behind the observed stimulated emission and we characterize the emission features for the proposed laser devices. Ionic Liquids (ILs) are presently defined as being organic salts that melt at or below 100° C [23]. Their importance evolves mainly from the thermo-physical and phase equilibrium properties, while the flexibility of their synthesis makes them perfect candidates as "designer materials" [24] for task explicit requirements. We present herein the experimental results for only a few selected dyes dissolved in ILs, showing the capacity to achieve laser emission from certain special configuration micrometric systems. We wish to stress that the discovery is generic and it can be broadened to roughly any IL and dye molecule [25].

The main schematic for the set-up used in the experiments is depicted in Figure 1. The pump laser is represented by a ns pulsed Nd:YAG laser (variable repetition rate 1-20Hz, pulse

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duration 5ns). The sample optical emission can be observed on a paper screen, as well as analyzed by means of a Charged Coupled Device (CCD) Ocean Optics spectrometer (resolution ca. 0.15nm), while the far field spatial profile measurements were performed with a Hi-Res 1,360 x 1,024 pixels 12bit CCD beam profiler camera (Thorlabs). For some of the experiments we substituted the classical spherical lens (f ca. 10cm) with an array of plano-convex spherical micro-lenses (Thorlabs; each micro lens has a diameter of ca. 150 microns).

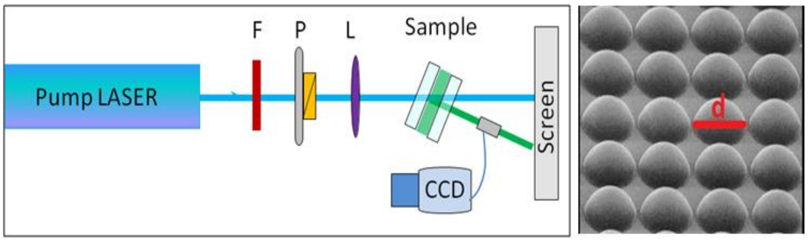

Fig. 1. (left) Experimental set-up for our study (F – an energy attenuator filter; P – optical polarizer; L – spherical lens; CCD – fiber Charged Coupled Device multichannel spectrometer). (right) Array of micro-lenses (from Thorlabs) used for creating a microstructured emission (d equals ca. 150 microns is the diameter of each plano-convex spherical micro-lens).

The samples were prepared by mixing ILs with ca. 0.3% by weight organic dyes. The measurements were carried out on the following two active mixtures: (1). Pyrromethene597 dye (4,4-Difluoro-2,6-di-t-butyl-1,3,5,7,8-pentamethyl-4-bora-3a,4a-diaza-s-indacene 2,6-Di-t-butyl-1,3,5,7,8-pentamethyl pyrromethene difluoroborate Complex) in Ionic Liquid 1-Butyl-3-methylimidazolium hexafluorophosphate; (2) DCM dye (4-Dicyanmethylene-2-methyl-6-(pdimethylaminostyryl)-4H -pyran) in Ionic Liquid Methyl-trioctylammoniumbis (trifluoromethylsulfonyl)imide). Samples consisted of glass wedge cells (variable thickness for the active matrix in the range 5-150 microns) and micrometric capillary glass tubes (diameter 10-200 microns).

2. Experimental results and discussions

A first experiment shows mirrorless random laser action from a glass capillary tube filled with mixture 1 (Figure 2(a)). Upon optical excitation with the Nd:YAG pump laser (at 532nm) we obtained highly intense laser emission. By means of a Thorlabs energy meter instrument we established the threshold value for this type of micro-laser device at around a few microJ/pulse (depending slightly also on the diameter of the capillary tube).

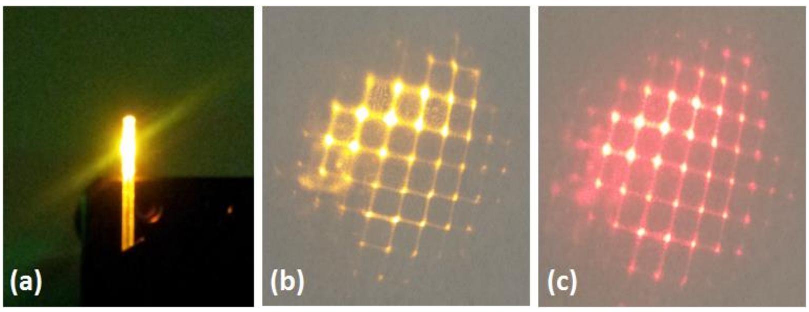

Fig. 2. (a) Picture of the lasing action from a cylindrical capillary sample. (b-c) Picture showing the On-Screen pattern of random laser micro-array emitters obtained by optically exciting wedge cell samples with the special micro-lenses system (Yellow (b) and Red (c) emissions are shown respectively).

When swapping the standard lens with the special micro-lens array from Fig. 1(b) we are able to obtain a very interesting scenario. While using enough input energy to pump a standard wedge cell filled with active mixture 1 (and then with active mixture 2) we can obtain a spectacular random laser micro-array emitter. In figure 2 (b) and 2 (c) one can observe a real picture of the emission coming from a yellow (mixture 1) micro-laser and a red (mixture 2) one. Please note that in this case we need higher pump energy to initiate the laser emission (optical losses due to the use of the micro-lens array are much higher; while also considering the Gaussian profile of the pump laser, it is understandable that more power overall is needed to "turn on" several micro lasers from the array when being slightly off the main Gaussian energy axis). We estimate to require at least 30-40 microJ/pulse (and up) to be able to initiate multiple micro-lasers from the array. Yet, the possibility to achieve such a beautiful structured emission is really remarkable and demonstrates once more the high efficiency of the random lasing active system.

The spectral measurements further demonstrate that lasing occurs in the case of our special micro-devices. Fig. 3 shows the emission spectrum in the case of a micro-cylindrical sample doped with mixture 1 (i.e. from Figure 2(a)). Laser action is confirmed at around 580nm wavelength when pumping above input energy threshold. The FWHM (Full Width Half Maximum) of the generated laser line is ca. 0.5nm. As expected in the case of random lasers, several adjacent laser modes may surface in the spectrum.

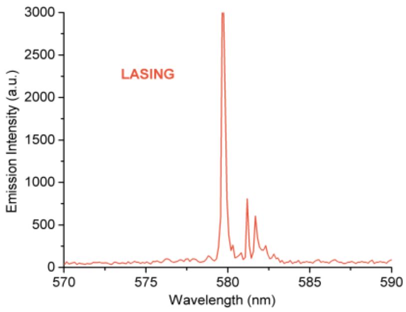

Fig. 3.Random lasing spectrum from the capillary sample (each lasing mode has a FWHM of ca. 0.5 nm)

Figure 4 shows instead the spectral characterization of the sample depicted in Figure 2(c). At low pump energy levels, one can achieve the typical wide spontaneous emission curve of the DCM dye used in the mixture, indicating that the IL host system does not significantly transform the fluorescence spectrum (Figure 4 (a)). The fluorescence spectrum is sharpened gradually (with increasing pump power) into ASE (Amplified Spontaneous Emission). A difference is clearly noticed when increasing the laser pump energy slightly above a given threshold value (about 30 microJ/pulse for this particular sample). Here (Figure 4(b)), we achieve sporadic mirrorless laser emission on top of the ASE curve. As soon as the incident pump energy further exceeds the threshold value, the lasing peaks intensities increase faster versus the pump intensity and also several sharper spikes appear, owing to the fact that, in this case, the balance between gain and loss of these lossiest modes become positive (note the narrow FWHM per lasing mode of less than 0.5nm) (Figure 4(c)).

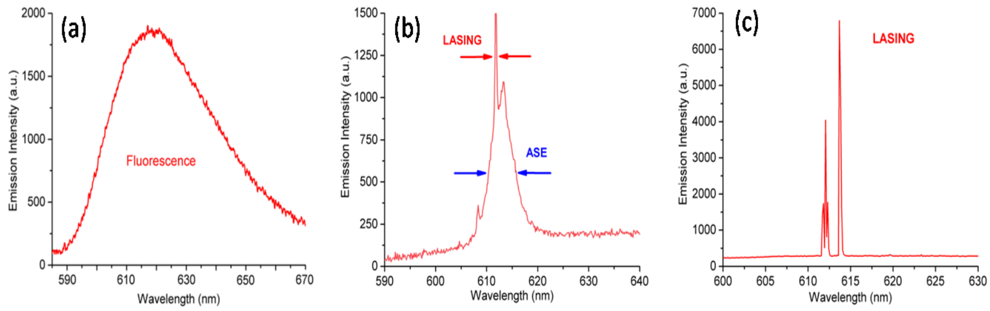

Fig. 4. Emission spectra from a wedge sample cell under micro-array pumping conditions for increasing input energies. (a) Regular fluorescence spectrum obtained for under-threshold pumping situation. (b) Spectrum acquired for near the threshold energy conditions (one can notice both ASE and emerging lasing peaks). (c) Spectrum for lasing in beyond energy threshold situation

Additional investigations that were performed in this study include the measurement of the far field spatial intensity profile of the samples in lasing conditions.

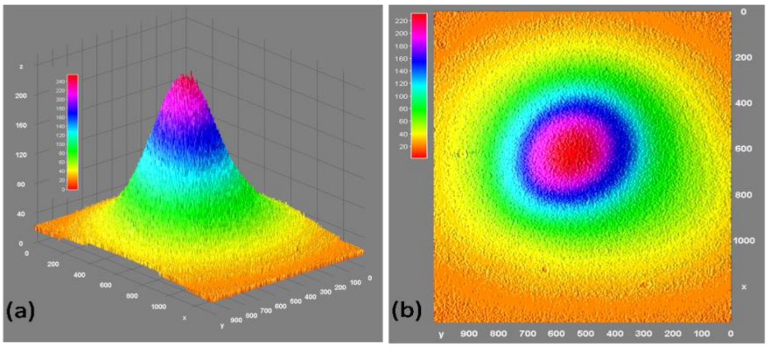

Fig. 5. Random lasing (one-shot) far field emission intensity profile from a micro-capillary in above threshold energy conditions. (a) 3D emission intensity profile. (b) 2D emission intensity profile

Fig. 5 depicts the lasing intensity profile from the sample in Fig. 2(a) acquired by means of a HiRes (1,360 x 1,024 pixels 12bit) CCD beam profiler camera. The overall shape for our lasing spot is somehow comparable to the pump laser Gaussian profile, as expected, when the system is situated above threshold energy conditions (Figure 5(a) and 5(b)). However, one can notice that this emission intensity profile is created by a sequence of bright spatially overlapped speckles that are generating a richly structured pattern. This feature is a typical characteristic of a random laser system.

Furthermore, we also acquired the emission intensity profile in High Resolution mode (over a very small area) for the laser action in case of the special micro-lens array configuration when using the slightly above the threshold energy situation (Figure 6). Again, as anticipated for a typical random laser behavior, the intensity profile displays a speckle like pattern (which can also be confirmed to vary for each pump pulse).

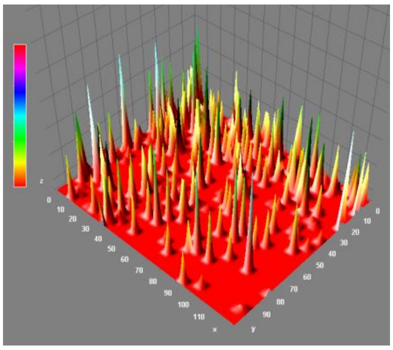

Fig. 6. High resolution random lasing (one-shot) 3D far field emission intensity profile (acquired from a wedge sample cell excited in the micro array configuration, in above threshold energy conditions).

We advance a hypothesis that a network of cavities is formed due to the salt nature of the solvent and the ability to solvate in different manner the more polar dyes with the smaller and more ionic liquid. Similar studies are present for various interactions in liquid crystalline materials [26,27]. Nevertheless, further investigations are required for better understanding the physics behind the development of these cavities. The estimated overall efficiency of the micro-capillary laser system (i.e. the output lasing energy divided by the input beam energy) is particularly high, nearly 30%, even when having a certain fraction from the incident beam not fully absorbed into the gain medium. A lower-grade situation is present in the case of the micro-lens array configuration (as explained before, in this case, the optical losses are higher). Nevertheless, we are convinced that these already great efficiency values can be additionally improved by optimizing various physical parameters of the systems and the set-up. Long term experiments revealed that the compact laser systems are highly reliable and robust (they maintain the emission properties even after extended optical pumping intervals).

3. Conclusions

We presented and characterized laser action phenomena in special micrometric configurations of dye doped ionic liquids confined in special geometries. Because of low vapor pressure, low inflammability, liquidity over a broad temperature range, recyclability these matrices are excellent contenders for building, as demonstrated, a series of interesting mirrorless laser devices. Cylindrical micro-capillaries and special micro-lens array systems grant the possibility to obtain highly-compact hazard-free narrow banded (FWHM ca. 0.5 nm for each laser mode) emitters that outshine because of the low laser threshold (a few microJ/pulse in some situations), superior efficiency, long term robustness, cheap construction costs and versatility in terms of potential configurations. A detailed spectral analysis for the designed samples and an interpretation of the far field modal profiles is performed in the paper, which clearly confirm a random laser behavior for these systems.

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